FORM PTO-1390 (Modified) REV 11-2000) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE 12075US03 TRANSMITTAL LETTER TO THE UNITED STATES U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR DESIGNATED/ELECTED OFFICE (DO/EO/US) 936465 CONCERNING A FILING UNDER 35 U.S.C. 371 INTERNATIONAL APPLICATION NO. PRIORITY DATE CLAIMED INTERNATIONAL FILING DATE PCT/US00/06254 10 March 2000 12 March 1999 TITLE OF INVENTION BEVERAGE CONTAINER CLOSURE AND SEALANT LAYER MATERIAL APPLICANT(S) FOR DO/EO/US NASEEM, Homaira and O'HARA, Charles J. Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371. 2. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include itens (5), (6), 3. (9) and (24) indicated below. 4. The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. \boxtimes A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) is attached hereto (required only if not communicated by the International Bureau). b. 🗆 has been communicated by the International Bureau. is not required, as the application was filed in the United States Receiving Office (RO/US). 6. П An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). is attached hereto. has been previously submitted under 35 U.S.C. 154(d)(4). 7. Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) П are attached hereto (required only if not communicated by the International Bureau). b. have been communicated by the International Bureau. c. have not been made; however, the time limit for making such amendments has NOT expired. d. 🗆 have not been made and will not be made. **-**8. An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). 10. An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). \boxtimes 11. A copy of the International Preliminary Examination Report (PCT/IPEA/409). 12. A copy of the International Search Report (PCT/ISA/210). Items 13 to 20 below concern document(s) or information included: 13. An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 14. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 15. A FIRST preliminary amendment. 16. A SECOND or SUBSEQUENT preliminary amendment. 17. A substitute specification. A change of power of attorney and/or address letter. 18. 19. A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 20. A second copy of the published international application under 35 U.S.C. 154(d)(4). A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 21. 22. \times Certificate of Mailing by Express Mail \boxtimes 23. Other items or information:

U.S. APPLICATION I	NO. (IF KNOWN, SEE 37 CFR 7 9 36 46 5	INTERNATIO APPLICATI PCT/US00/0625			OOCKET NUMBER 5US03
24. The following	lowing fees are submitted:.			CALCULATIONS	PTO USE ONLY
BASIC NATIONA	L FEE (37 CFR 1.492 (a) (1) -		j		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO					
	 ✓ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO				
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☐ International but all claim	☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)				
☐ International and all claim	I preliminary examination fee (37 as satisfied provisions of PCT Art	CFR 1.482) paid to USPTO icle 33(1)-(4)	\$100.00		
	ENTER APPROPRI	ATE BASIC FEE AMO	OUNT =	\$860.00	
Surcharge of \$130.0 months from the ear	00 for furnishing the oath or declar liest claimed priority date (37 Cl	ration later than \Box 20 FR 1.492 (e)).) □ 30	\$0.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	13 - 20 =	0	x \$18.00	\$0.00	
Independent claims	3 - 3=	0	x \$80.00	\$0.00	
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reduced by 1/2	ms small entity status. (See 37 CF	R 1.27). The fees indicated above	ve are	\$0.00	
		SUB	ΓOTAL =	\$860.00	
Processing fee of \$1 months from the ear	30.00 for furnishing the English cliest claimed priority date (37 CI	translation later than \Box 20 FR 1.492 (f)).	O □ 30 +	\$0.00	
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Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).					
TOTAL FEES ENCLOSED = \$860.00					
				Amount to be:	\$
unit.				refunded charged	\$
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b. 🗌 Plea					
c. 🗵 The					
to Deposit Account No. 13-0017 A duplicate copy of this sheet is enclosed. d. Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card					
information should not be included on this form. Provide credit card information and authorization on PTO-2038.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO:					
SIGNATURE					
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McAndrews, Held & Malloy, Ltd. 500 W. Madison Street Robert W. I				esler ———————	
Suite 3400 NAME					
Chicago, Illinois 60661 Tel: 312/775-8000 Reg. No. 31			Reg. No. 31,8	26	
Fax: 312/775-8100			REGISTRATIO	N NUMBER	
September 12, 2001					
K			DATE		

BEVERAGE CONTAINER CLOSURE AND SEALANT LAYER MATERIAL

Field Of The Invention

The present invention relates to thermoplastic materials used to mold beverage container closures and sealant layers. More particularly, the present invention relates to thermoplastic materials having improved gas barrier properties.

Background Of The Invention

Beverage containers formed from moldable thermoplastic such as polyethylene terephthalate generally have a threaded opening that accommodates a threaded closure or cap. The closure is molded from thermoplastic materials such as polypropylene homopolymer, polypropylene copolymer, and high density polyethylene. A sealant layer or liner is generally interposed between the closure and the beverage container to prevent fluid leakage from or into the container.

An important property of beverage container closure and sealant layer materials is the barrier or permeability of the materials. In this regard, traditional polypropylene and polyethylene closure and sealant materials were developed without primary regard for their gas barrier properties. More recently, efforts have been made to develop improved closure and sealant materials having lower permeability to oxygen and carbon dioxide. Such improved materials would be preferred for beverage containers, especially carbonated beverage containers, for which extended shelf life and beverage quality is demanded.

It has been found that the addition of

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nanocomposite, montmorillonite clay to thermoplastic materials used in the molding of beverage container closures and sealant layers improves the barrier properties of the materials. Specifically, the addition of montmorillonite clay decreases the permeability of such thermoplastic materials to oxygen and carbon dioxide.

Summary Of The Invention

A melt-processible composition for molding closures for beverage containers has improved gas barrier properties. The composition comprises:

- a thermoplastic base polymeric material;
- a quantity of layered magnesium aluminum silicate clay having platelets with a diameter of approximately 1 micron.

In the preferred closure composition, the thermoplastic base polymeric material comprises a polyolefin such as polypropylene, polyethylene and/or a copolymer comprising propylene and ethylene monomeric units.

The layered magnesium aluminum silicate clay is preferably a nanocomposite, montmorillonite clay.

A melt-processible composition for molding sealant layers for beverage containers also has improved gas barrier properties. The composition comprises:

- (a) a thermoplastic base polymeric material;
- a quantity of layered magnesium aluminum silicate clay having platelets with a diameter of approximately 1 micron.

In the preferred sealant layer composition, the thermoplastic base polymeric material comprises ethyene

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vinyl acetate copolymer, polyethylene (linear low density polyethylene, low density polyethylene, ultra-low density polyethylene, and high density polyethylene), styrene ethylene butadiene styrene polymer (commercially available from Shell under the trade designation KRATON®), styrene butadiene styrene polymer, ethylene propylene diene monomer, and metallocene polymers. The layered magnesium aluminum silicate clay is preferably nanocomposite, montmorillonite clay.

A method of decreasing the gas permeability of a thermoplastic material comprises introducing a quantity of layered magnesium alumnium silicate clay into the material. The preferred magnesium aluminum silicate clay is nanocomposite, montmorillonite clay.

Detailed Description Of The Preferred Embodiments

Polypropylene-based thermoplastic compositions, suitable for molding beverage container closures, were prepared using the following formulations:

Sample No. 98-006 control:

<u>Material</u>	Chemical Composition	Parts by Weight
Montell CA 012	Polypropylene copolymer	97
5019466JEMB	White color masterbatch	3

<u>Sample No. 98-006 B</u>:

<u>Material</u>	Chemical Composition	Parts by Weight
Montell CA 012	Polypropylene copolymer	95
5019466JEMB	White color masterbatch	3
Cloisite 25 A	Nanocomposite clay	3 to 10

The oxygen permeability for plaques (30 mils nominal thickness) of each of the above samples was measured on a Mocon Ox-Tran 10/50A analyzer at $23.2^{\circ}C$ and 0% relative

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humidity. The system was purged for 2 hours and each plaque was conditioned in a 100% oxygen atmosphere overnight prior to testing. Test area was 50 cm². Oxygen permeability was measured on three plaques for each sample and the results were averaged and reported below:

Sample		<u>Oxygen</u>	Permeability	(cc/m²/day)
98-006	control		110.8	
98-006	В		90.6	

The above results showed that the addition of nanocomposite, montmorillonite clay to the polypropylene copolymeric thermoplastic base material decreased the oxygen permeability of the material by approximately 18%.

Ethylene-vinyl acetate (EVA) thermoplastic compositions, suitable for molding beverage container liners, were prepared using the following formulations:

Sample No. 240-6C control:

<u>Material</u>	Chemical Composition	Parts by Weight
UE 655	9% EVA 2 melt index	15
UE 635	9% EVA 9.8 melt index	85
Escorene 3505G	Polypropylene homopolyme	er 5
	400 melt index	
Crodamine ER	Erucamide	1
Aldo MS	Glycerol monostearate	0.1

Sample No. 97-692:

	<u>Material</u>	Chemical Composition Par	ts by Weight
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	UE 655	9% EVA 2 melt index	5 to 15
	UE 635	9% EVA 9.8 melt index	80 to 100
	Escorene 3505G	Polypropylene homopolymer	3 to 10
		400 melt index	
35	Crodamine ER	Erucamide	0.5 to 2.0

Cloisite 25 A Nanocomposite clay 3 to 10 Aldo MS 0.1 to 0.5 Glycerol monostearate

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The oxygen permeability for plaques (30 mils nominal thickness) of each of the above samples was measured on a Mocon Ox-Tran 10/50A analyzer at 23.2°C and 0% relative humidity. The system was purged for 2 hours and each plaque was conditioned in a 100% oxygen atmosphere overnight prior to testing. Test area was 50 cm2. Oxygen permeability was measured on three plaques for each sample and the results were averaged and reported below:

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240-6C control

Oxygen Permeability (cc/m²/day) 343.8

97-692

Sample

302.2

The above results showed that the addition of montmorillonite clay to the EVA thermoplastic base material decreased the oxygen permeability by approximately 12%.

The permeability to carbon dioxide, as measured in % gas loss/day, improved approximately 15% (i.e., the % gas loss/day decreased approximately 15%) with the addition of nanocomposite, montmorillonite clay.

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A melt-processible composition for molding closures for beverage containers comprising:

- a thermoplastic base polymeric material;
- (b) a quantity of layered magnesium aluminum silicate clay having platelets with a diameter of approximately 1 micron.

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2. The composition of claim 1 wherein said thermoplastic base polymeric material comprises a polyolefin.

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3. The composition of claim 2 wherein said polyolefin is selected from the group consisting of polypropylene, polyethylene and a copolymer comprising propylene and ethylene monomeric units.

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The composition of claim 1 wherein said layered magnesium aluminum silicate clay is montmorillonite clay.

- A beverage container closure molded from the composition of claim 1.
- 25 A melt-processible composition for molding 6. sealant layers for beverage containers comprising:
 - (a) a thermoplastic base polymeric material;
 - a quantity of layered magnesium aluminum silicate clay having platelets with a diameter of approximately 1 micron.

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7. The composition of claim 7 wherein said

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thermoplastic base polymeric material is selected from the group consisting of ethyene vinyl acetate copolymer, polyethylene, styrene ethylene butadiene styrene polymer, styrene butadiene styrene polymer, ethylene propylene diene monomer, and metallocene polymers.

- 8. The composition of claim 6 wherein said layered magnesium aluminum silicate clay is montmorillonite clay.
- 9. A beverage container sealant layer molded from the composition of claim 6.
- 10. A method of decreasing the gas permeability of a thermoplastic material, said method comprising introducing a quantity of layered magnesium aluminum silicate clay to said material.
- 11. The method of claim 10 wherein said thermoplastic material is a polyolefin.
- 12. The method of claim 11 wherein said polyolefin is selected from the group consisting of polypropylene, polyethylene and a copolymer comprising propylene and ethylene monomeric units.
- 13. The method of claim 10 wherein said layered magnesium aluminum silicate clay is montmorillonite clay.



DECLARATION AND POWER OF ATTORNEY

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe that I am the original inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

BEVERAGE CONTAINER CLOSURE AND SEALANT LAYER MATERIAL WITH IMPROVED BARRIER PROPERTIES

the specification of which was filed in the U.S. Patent and Trademark Office on December 19, 2001 (international filing date March 10, 2000), and assigned Serial No. 09/936,465.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims. I acknowledge the duty to disclose information that is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, Section 1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, section 119(a)-(d) or section 365(b) of any foreign application(s) for patent or inventor's certificate, or section 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application on which priority is claimed:

International Application No. PCT/USOO/06254 filed March 10, 2000.

I hereby claim the benefit under Title 35, United States Code, section 119(e) of any United States provisional application(s) listed below:

U.S. Provisional Patent Application Serial No. 60/124,102 filed March 12, 1999.

I hereby claim the benefit under Title 35, United States Code, section 120 of any United States application(s), or section 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in such prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, section 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, section 1.56(a) which became available between the filing date of the prior application(s) and the national or PCT international filing date of this application:

NONE.

I hereby appoint:



George P. McAndrews Reg. No. 22,760 John J. Held Reg. No. 21,061 Timothy J. Malloy Reg. No. 25,600 William M. Wesley Reg. No. 26,521 Lawrence M. Jarvis Reg. No. 27,341 Gregory J. Vogler Reg. No 31,313 Jean Dudek Kuelper Reg. No. 30,171 Herbert D. Hart III Reg. No. 30,063 Robert W. Fieseler Reg. No. 31,826 Thomas J. Wimbiscus Reg. No. 36,059 Steven J. Hampton Reg. No. 33,707 Reg. No. 32,223 Reg. No. 30,590 Priscilla F. Gallagher Stephen F. Sherry Patrick J. Arnold Jr. Reg. No. 37,769 George Wheeler Reg. No. 28,766 Ronald E. Larson Reg. No. 24,478 Christopher C. Winslade Reg. No. 36,308 Edward A. Mas II Reg. No. 37,179 Gregory C. Schodde Reg. No. 36,668 Edward W. Remus Reg. No. 25,703 Donald J. Pochopien Reg. No. 32,167 Sharon A. Hwang Reg. No. 39,717 David D. Headrick Reg. No. 40,642 Dean D. Small Reg. No. 34,730 Alejandro Menchaca Reg. No. 34,389 Kirk A. Vander Leest Reg. No. 34,036

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the address and telephone number of each of whom is McAndrews, Held & Malloy, Ltd., 500 West Madison Street, 34th Floor, Chicago, Illinois 60661, (312) 775-8000, as our attorneys with full power of substitution and revocation to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

Please direct telephone calls to Robert W. Fieseler at (312) 775-8123. Please direct facsimiles to Robert W. Fieseler at (312) 775-8100.

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Robert W. Fieseler McAndrews, Held & Malloy, Ltd. 500 West Madison Street, Suite 3400 Chicago, Illinois 60661 I hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

1-D	O .	
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